Carbone *et al.* Reply: In [1], the deflection of an electron beam passing close to a graphite surface photoexcited by infrared femtosecond laser pulses is reported. The authors claim that such deflection could contribute to the observations in [2]. Neither the dependence on diffraction orders and intensity [3] nor the strength and time scale of the transient electric fields (TEF) induced deflection can justify the claim. The following details illustrate the point.

(i) The dynamics of different Bragg diffraction orders obey diffraction laws [3]; see Figs. 1(a) and 1(b). This only occurs if the dynamics are structural in origin.

(ii) TEF effects have been modeled theoretically and their influence on Bragg dynamics has been investigated experimentally in [3] in a quantitative way; they have been shown to be a small effect compared to structural dynamics, especially for low excitation fluences.

(iii) By comparing the dynamics of the diffracted beam in [2] to the deflections induced by TEF reported in [1] in the same geometrical conditions, we show that this deflection is small compared to the structural motions discussed in [2]. In particular, for low excitation fluence, which was the focus of [2], hardly any deflection was observed in [1].

(iv) The effect reported in [2], i.e., the compression of graphite lattice upon laser irradiation, has been confirmed by different independent ultrafast electron diffraction experiments [4,5], experiments with different techniques including ultrafast electron microscopy in transmission through a graphite thin film [6,7], and experiments not involving electrons as a probe like STM [8].

Clearly, if the fluence is very high one must consider the electrons ejected from the material and the field they may produce. In order to illustrate the point, we redraw the Bragg peaks motions reported in [2] in angular deflections; see Figs. 1(c) and 1(d) (the precise experimental geometry is described in [10]). In the same figure we plot the data taken from [1] for the low fluence and the high fluence regime. In Fig. 1(c), we compare the Bragg peak position change to the beam deflection by TEF at high fluence ($44\ \mathrm{mJ/cm^2}$); for a sample to beam distance of 92 μm, which is the smallest value reported in Fig. 1(a) of [1], the deflection is the strongest. In this case, while TEF could influence the long-term dynamics, its effect in the 0–10 ps time scale, where the compression was observed in [2], is minimal. In Fig. 1(d), we compare the Bragg peak dynamics to the deflection induced by TEF for low excitation fluence ($7\ \mathrm{mJ/cm^2}$). In this case, the data reported in [1] show basically no effect, either on the long-time scale or on the short-time scale, while in [2] the ultrafast compression was the clearest in the low fluence regime. In conclusion, TEF could affect the motion of Bragg peaks in the long-time dynamics and for high pump fluences; however, the critical control tests are clear: the diffraction order dependence, the intensity changes with fluence, and the comparison with the direct beam behavior [3] confirm that the dynamics reported in [2] are structural in origin. As shown in [3], the effect is irrelevant in ultrafast electron microscopy, contrary to the claim made [9], and the results for graphite are also evident in these microscopy studies.

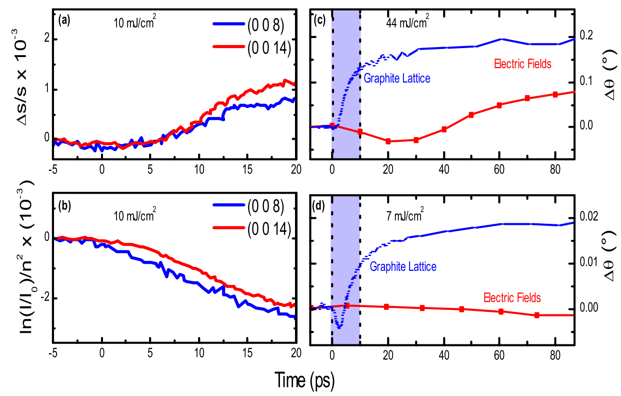

FIG. 1 (color online). (a) Normalized changes in the scattering vector $\Delta s/s$ for the Bragg spots (0 0 8), (0 0 14). The changes of the diffracted beams are linear with the diffraction order, whereas the rise times do not depend on s, as expected for structural dynamics (data adapted from [3]). (b) Temporal evolution of the Bragg spot intensity, consistent with the Debye-Waller s^2 dependence on the diffraction order. (c),(d) Dynamics of the Bragg peak together with deflections at low fluence ($7\ \mathrm{mJ/cm^2}$), and high fluence ($44\ \mathrm{mJ/cm^2}$), where a shift of 15 pixels of the (0 0 14) order was recorded on a CCD camera 17 cm away (pixel size 45 μm [10]), corresponding to a maximum deflection of $2\theta = 0.23°$ and a scattering angle $\theta = 0.11°$, and corresponding to a change around 9 pm of the interlayer distance of graphite, and of 18 pm of the whole unit cell.

Fabrizio Carbone,[1] Peter Baum,[1] Petra Rudolf,[2] and Ahmed H. Zewail[1]
[1]Physical Biology Center for Ultrafast Science and Technology
Arthur Amos Noyes Laboratory of Chemical Physics
California Institute of Technology
Pasadena, California, 91125, USA
[2]Zernike Institute for Advanced Materials
University of Groningen, Nijenborgh 4,
NL-9747AG Groningen, The Netherlands

Received 7 June 2010; published 30 July 2010
DOI: 10.1103/PhysRevLett.105.059604
PACS numbers: 61.82.−d, 07.78.+s, 63.20.K−, 78.47.−p

[1] H. Park and J. M. Zuo, preceding comment, Phys. Rev. Lett. **105**, 059603 (2010).
[2] F. Carbone *et al.*, Phys. Rev. Lett. **100**, 035501 (2008).
[3] S. Schäfer, W. Liang, and A. H. Zewail, Chem. Phys. Lett. **493**, 11 (2010).
[4] R. K. Raman *et al.*, Phys. Rev. Lett. **101**, 077401 (2008).
[5] C.-Y. Ruan *et al.*, Nano Lett. **7**, 1290 (2007).
[6] F. Carbone *et al.*, Chem. Phys. Lett. **468**, 107 (2009).
[7] F. Carbone, O. H. Kwon, and A. H. Zewail, Science **325**, 181 (2009).
[8] J. Kanasaki *et al.*, Phys. Rev. Lett. **102**, 087402 (2009).
[9] H. Park and J. M. Zuo, Appl. Phys. Lett. **94**, 251103 (2009).
[10] D.-S. Yang *et al.*, J. Phys. Chem. C **111**, 4889 (2007).

